

In the Specification

On page 1, kindly replace the first paragraph with the following:

TECHNICAL FIELD

The present invention relates to thermoplastic resin structures of good vapor and/or liquid transmission resistance, and their use. In particular, the invention relates to resin moldings of specific transmission resistance, low water absorption, dimensional stability in ~~wet~~moisture and workability, which are obtained by forming a specific morphology of polyamide resin and polyphenylene sulfide resin and are favorable for vapor and/or liquid barrier articles, to multi-layer structures favorable for containers and pipes for storage and transportation of liquid chemicals such as oil and gasoline, to those favorable for wrapping and packaging materials and containers for foods and medicines, and to their use.

Kindly replace the paragraph spanning pages 2 and 3 with the following:

The method could improve the dimensional stability and the stiffness of the resin compositions and moldings in ~~wet~~moisture over that of moldings of polyamide resin alone, but is still unsatisfactory. When the resin compositions and moldings are used for structures that are required to have good transmission resistance and toughness, they are not satisfactory. Therefore desired are molding materials that comprise polyamide resin and PPS resin and have both the good properties of polyamide resin and the good properties of PPS resin in good balance.

Kindly replace the paragraph spanning pages 3 and 4 with the following:

The present invention is to realize a high-level balance of the mechanical strength and the toughness of polyamide resin and the low water absorption and the transmission resistance of PPS resin, and its object is to provide thermoplastic resin structures which are no improved to the highest possible degree that they are free from the reduction in the mechanical properties such as dimen-

sional stability and stiffness and the reduction in the liquid chemical and vapor transmission resistance intrinsic to polyamide resin in wetmoisture, especially to provide polyamide-PPS resin moldings favorable for vapor and/or liquid barrier articles, and to provide multi-layer structures of good transmission resistance, moldability, workability, interlayer adhesiveness and toughness that are favorable to plastic containers and can be stably and economically formed into good plastic containers.

Kindly replace the paragraph spanning pages 11 and 12 with the following:

Preferably, the polyamide resin for use in the invention contains a copper compound for improving the long-term heat resistance thereof. Examples of the copper compound are cuprous chloride, cupric chloride, cuprous bromide, cupric bromide, cuprous iodide, cupric iodide, cupric sulfate, cupric nitrate, copper phosphate, cuprous acetate, cupric acetate, cupric salicylate, cupric stearate, cupric benzoate, and chelate compounds of such an inorganic copper halide with xylylene-diamine, 2-mercaptobenzimidazole or benzimidazole. Especially preferred are cuprous compounds, and more preferred are cuprous halides. Examples of the preferred copper compounds are cuprous acetate and cuprous iodide. The amount of the copper compound to be added to the polyamide resin preferably falls between 0.01 and 2 parts by weight, more preferably between 0.015 and 1 part by weight, relative to 100 parts by weight of the polyamide resin. If there is too much copper compound, the copper compound will release metal copper while the resin composition is molded in melt, and will discolor the moldings. The commercial value of discolored moldings is low. Combined with such a copper compound, an alkali halide may also be added to the polyamide resin. Examples of the alkali halide are lithium chloride, lithium bromide, lithium iodide, potassium chloride, potassium bromide, potassium iodide, sodium bromide, and sodium iodide. Especially preferred are potassium iodide and sodium iodide.

Kindly replace the paragraph spanning pages 20 and 21 with the following:

The blend ratio of the polyamide resin (a) and the PPS resin (b) that constitute the thermoplastic resin structure of the invention is described. In the case where the PPS resin component forms a continuous phase (matrix phase) and the polyamide resin component forms a disperse phase in the morphology of the structure (for example, like a sea-island configuration as in Fig. 1), the polyamide resin accounts for from 5 to 80 % by volume and the PPS resin for from 20 to 95 % by volume. Preferably, the polyamide resin accounts for from 55 to 90 % by volume and the PPS resin for from 20 to 45 % by volume. In that case where the amount of the PPS resin component is small, the morphology in which the PPS resin forms a continuous phase can be formed, for example, by suitably controlling the melt viscosity ratio of polyamide resin/PPS resin. The moldings having the morphology have a good balance of wet characteristics and transmission resistance and, when they are used for the barrier layer in multi-layer structures, they also have a good balance of toughness, interlayer adhesiveness, barrier properties and cost. Therefore, they are extremely favorable. More preferably, the blend ratio of the two components is such that the polyamide resin accounts for from 60 to 75 % by volume and the PPS resin for from 25 to 40 % by volume. If the polyamide resin component (a) is larger than 80 % by volume, the PPS resin component could hardly form the continuous phase characteristic of the resin moldings of the invention, and the object of the invention cannot be attained. On the other hand, if the polyamide resin component (a) is smaller than 5 % by volume, it is unfavorable since the resin moldings could not be tough and the multi-layer structures could not have good interlayer adhesiveness.

Kindly replace the paragraph spanning pages 21 and 22 with the following:

In the case where the PPS resin component and the polyamide resin component both form substantially continuous phases (matrix phases) in the morphology of the resin structure (for

example, like a sea-sea configuration as in Fig. 2), it is important that the melt viscosity and the compatibility of the polyamide resin and the PPS resin are controlled within a composition range of such that the polyamide resin accounts for from 15 to 85 % by volume and the PPS resin for from 15 to 85 % by volume. For embodying the separated morphology in that manner, the blend ratio of the two components is preferably such that the polyamide resin accounts for from 30 to 70 % by volume and the PPS resin for from 30 to 70 % by volume, more preferably such that the polyamide resin accounts for from 35 to 65 % by volume and the PPS resin for from 35 to 65 % by volume. If the polyamide resin component (a) is larger than 85 % by volume, the PPS resin component could hardly form a substantially continuous phase, and structures that attain the object of the invention could not be obtained.

On page 40, kindly replace the third paragraph with the following:

(2) Alcohol gasoline transmission in wetmoisture:

The test tube filled with the alcohol gasoline mixture in the same manner as in the above (1) was kept in a thermo-hygrostat at 40°C and 65 % RH for 500 hours, and its weight loss was measured.

On page 41, kindly replace the third and fourth full paragraphs with the following:

(6) Dimensional stability in wetmoisture:

The test pieces were wetted in the same manner as in the test for water absorption. The length increase in wetmoisture was obtained from the length (in the machine direction) of the dry test piece just after prepared and the length of the wetted test piece.

Dimensional stability in wetmoisture (%) = {(length of wetted test piece – length of dry test piece)/(length of dry test piece)} × 100.

Kindly replace the paragraph spanning pages 41 and 42 with the following:

(7) Flexural modulus in ~~wet~~moisture:

The flexural modulus of the test piece wetted in the same manner as in the test for water absorption was measured according to ASTM D790.

On page 46, kindly replace the first full paragraph with the following:

Examples 1 to 12, Comparative Examples 1 to 4:

As in Tables 1 and 2, the polyamide resin and the PPS resin were mixed and fed into a double-screw extruder, Nippon Seikoshō's TEX 30 Model through its main feeder. The inorganic filler, if used, was fed thereinto through a side feeder provided at some part of the cylinder. These were kneaded ~~in-melt~~and heated in the extruder at 300°C, for which the screw revolution was 200 rpm. The resulting pellets were dried, and then injection-molded into test pieces. The injection-molding machine used was Toshiba Kikai's IS100FA, and the mold temperature was 80°C. In addition, the pellets prepared in the same manner as above were molded into tubes for the test for alcohol gasoline transmission through them. The data of the transmission resistance, the mechanical strength and the physical properties in ~~wet~~moisture of the samples are given in Tables 1 and 2.

On page 48, kindly replace Table 1 with the following:

Table 1

| | Item | Unit | Ex. 1 | Ex. 2 | Ex. 3 | Ex. 4 | Ex. 5 | Ex. 6 | Co. Ex. 1 | Co. Ex. 2 |
|---------------------------|---|------------------|-----------|-----------|------------------------|-----------|-----------|-----------|-----------|--------------------------|
| Constituent Components | Type of polyamide resin amount | - vol. % | N6-2 70 | N6-2 60 | N6-2 90 | N66 70 | N6/66 70 | N6-2 35 | N6-1 100 | N6-1 70 |
| | Type of PPS resin amount | - vol. % | PPS-1 30 | PPS-2 40 | PPS-1 10 | PPS-1 30 | PPS-1 30 | PPS-2 65 | - | PPS-2 30 |
| | Polyamide/PPS melt viscosity ratio (kneading temperature, °C) | | 5.5 (300) | 3.6 (300) | 5.5 (300) | 4.7 (300) | 1.8 (300) | 3.6 (300) | - | 0.6 (300) |
| Separated Morphology | | continuous phase | PPS | PPS & PA | PA | PPS | PPS & PA | PPS & PA | PA | PA |
| | | disperse phase | PA | - | PPS laminar dispersion | PA | - | - | - | PPS spherical dispersion |
| Transmission Resistance | Alcohol Gasoline Transmission | g | 0.2 | 0.4 | 0.6 | 0.3 | 0.4 | 0.1 | 1.7 | 1.5 |
| | Alcohol Gasoline Transmission in wetmoisture | g | 0.4 | 0.8 | 1.1 | 0.7 | 0.6 | 0.3 | 1.9 | 1.6 |
| | Oxygen Transmission | note 1 | 10 | 10 | 25 | 10 | 20 | 5 | 50 | 45 |
| Mechanical Strength | Tensile Strength | MPa | 70 | 75 | 82 | 84 | 73 | 71 | 72 | 66 |
| | Flexural Modulus | GPa | 2.9 | 3.0 | 3.3 | 2.9 | 2.8 | 3.1 | 2.9 | 2.8 |
| | Izod Impact Strength | J/m | 49 | 45 | 55 | 51 | 52 | 43 | 54 | 28 |
| Properties in wetmoisture | Water Absorption | % | 1.3 | 0.9 | 3.0 | 1.2 | 1.4 | 0.3 | 3.9 | 2.1 |
| | Dimensional Stability | % | 0.04 | 0.06 | 0.65 | 0.05 | 0.06 | 0.01 | 0.58 | 0.19 |
| | Flexural Modulus | GPa | 1.7 | 2.0 | 1.1 | 1.8 | 1.6 | 2.2 | 0.6 | 1.1 |

- (N6-1): nylon 6 resin with m.p. 225°C and relative viscosity 2.80.
(N6-2): nylon 6 resin with m.p. 225°C and relative viscosity 3.30
(N66): nylon 66 resin with m.p. 265°C and relative viscosity 3.20.
(N6/66): nylon 6/66 copolymer with m.p. 217°C and relative viscosity 2.85.
(PPS-1): PPS resin with m.p. 280°C, MFR 1000 g/10 min, and Mw 30000.
(PPS-2): PPS resin with m.p. 280°C, MFR 300 g/10 min, and Mw 49000.

On page 49, kindly replace Table 2 with the following:

Table 2

| | Item | Unit | Ex. 7 | Ex. 8 | Ex. 9 | Ex. 10 | Ex. 11 | Ex. 12 | Co. Ex. 3 | Co. Ex. 4 |
|----------------------------------|---|------------------|-----------|-----------|------------------------|--------------|-------------|-----------|-----------|--------------------------|
| Constituent Components | Type of polyamide resin amount | - vol.% | N6-2 70 | N6-2 60 | N6-1 75 | N66 70 | N6/66 70 | N6-2 35 | N6-1 100 | N6-1 70 |
| | Type of PPS resin amount | - vol.% | PPS-1 30 | PPS-2 40 | PPS-1 25 | PPS-1 30 | PPS-1 30 | PPS-2 65 | - | PPS-2 30 |
| | Type of inorganic Filler amount | - vol.% | GF 40 | GF 40 | GF 40 | GF/talc 35/5 | GF/MF 30/10 | GF 40 | GF 40 | GF 40 |
| | Polyamide/PPS melt viscosity ratio (kneading temperature, °C) | | 5.5 (300) | 3.6 (300) | 1.6 (300) | 4.7 (300) | 1.8 (300) | 3.6 (300) | - | 0.6 (300) |
| Separated Morphology | | continuous phase | PPS | PPS & PA | PA | PPS | PPS & PA | PPS & PA | PA | PA |
| | | cisperse phase | PA | - | PPS laminar dispersion | PA | - | - | - | PPS spherical dispersion |
| Transmission Resistance | Alcohol Gasoline Transmission | g | 0.2 | 0.3 | 0.3 | 0.3 | 0.3 | 0.1 | 1.5 | 1.4 |
| | Alcohol Gasoline Transmission in <u>wetmoisture</u> | g | 0.5 | 0.7 | 0.7 | 0.6 | 0.6 | 0.3 | 1.7 | 1.6 |
| | Oxygen Transmission | note 1 | 10 | 10 | 15 | 10 | 20 | 5 | 50 | 45 |
| Mechanical Strength | Tensile Strength | MPa | 165 | 150 | 175 | 170 | 165 | 135 | 185 | 160 |
| | Flexural Modulus | GPa | 9.2 | 9.5 | 9.0 | 9.3 | 9.0 | 9.5 | 8.5 | 9.0 |
| | Izod Impact Strength | J/m | 100 | 90 | 110 | 100 | 105 | 90 | 110 | 85 |
| Properties in <u>Wetmoisture</u> | Water Absorption | % | 0.7 | 0.6 | 0.9 | 0.6 | 0.7 | 0.3 | 3.0 | 1.5 |
| | Dimensional Stability | % | 0.03 | 0.02 | 0.03 | 0.03 | 0.03 | 0.01 | 0.13 | 0.05 |
| | Flexural Modulus | GPa | 7.0 | 7.4 | 6.5 | 7.3 | 6.9 | 8.6 | 4.0 | 5.5 |

- (N6-1): nylon 6 resin with m.p. 225°C and relative viscosity 2.80.
(N6-2): nylon 6 resin with m.p. 225°C and relative viscosity 3.30
(N66): nylon 66 resin with m.p. 265°C and relative viscosity 3.20.
(N6/66): nylon 6/66 copolymer with m.p. 217°C and relative viscosity 2.85.
(PPS-1): PPS resin with m.p. 280°C, MFR 1000 g/10 min, and Mw 30000.
(PPS-2): PPS resin with m.p. 280°C, MFR 300 g/10 min, and Mw 49000.